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High Resolution Color Organic Light Emitting Diode Microdisplay Fabrication Method

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ABSTRACT

We report experimental results on the evaluation of a sealing technique for organic light emitting diodes (OLEDs) used for high-resolution color microdisplay applications. Based on production and processing requirements for active matrix OLEDs (AMOLEDs) on silicon, the sealing process must protect the device against moisture and oxygen, not only during operation and storage but also during production. A conformal polymer/metal(oxide)/polymer multi-layer technology was selected for this purpose. AMOLED test structures were produced and tested with and without sealing under ambient conditions as well as in water. The applied sealing process was shown to be compatible with all steps of the OLED-based microdisplay production of eMagin Corporation. Our results confirm that encapsulating AMOLEDs with the described process leads to increased stability, both under operation as well as for storage purposes.

Key Words: Organic Light Emitting Diodes, Active Matrix, Microdisplays, Sealing.

1. INTRODUCTION

Microdisplays are typically sized less than 2 inches diagonal, and viewed with magnifying optics. When properly combined, integrated circuits, microdisplays, and optics create a virtual image similar to the real image of a computer monitor or large screen TV. These high-performance, virtual imaging modules provide access to information-rich text, data, and video which can facilitate the opening of new mass markets for wearable PCs, wireless Internet appliances and mobile phones, portable DVD-viewers, digital cameras, and other emerging applications. There are several technologies used for microdisplays such as (1) digital micromirrors[1], (2) electrostatic microshutter arrays[2], (3) microdisplays using switchable Bragg grating technology[3], (4) liquid crystal displays (LCDs) placed over a uniformly illuminated backlight and (5) emissive microdisplays.

eMagin's microdisplays are emissive, based on a combination of fundamental OLED (organic light emitting diode) technology originally developed by Eastman Kodak and eMagin's technology. The electro-optical information transfer in OLEDs can be done via the process of controllable light emission as opposed to LCDs which use controllable light absorption. Hence LCDs require a separate light source and their efficiency is primarily controlled by the efficiency of this secondary light source. The overall efficiency for LCD-based systems is further decreased by the fact that they have to produce and re-absorb light in order to display dark areas. Hence emission-based displays use less power and are capable of high brightness and color. Emissive displays have one further general advantage: because the light they emit is "Lambertian" (i.e., appears equally bright from all directions), they are ideal for near to the eye applications since a small movement in the eye does not change the image, making virtual images appear more natural.

The choice of organic materials as the emitter is made because compared to inorganic EL materials the voltage is lower and the efficiency higher. Many organic materials with very different distinct charge transport and light emission properties are already available and being developed. This allows one to control the spectral and spatial region and efficiency of light

emission. Emission follows electron-hole recombination and singlet exciton formation. However, there is a trade off: the ease of production is coupled to a semiconductor layer which is very susceptible to external influences which could potentially reduce the lifetime of the OLEDs.

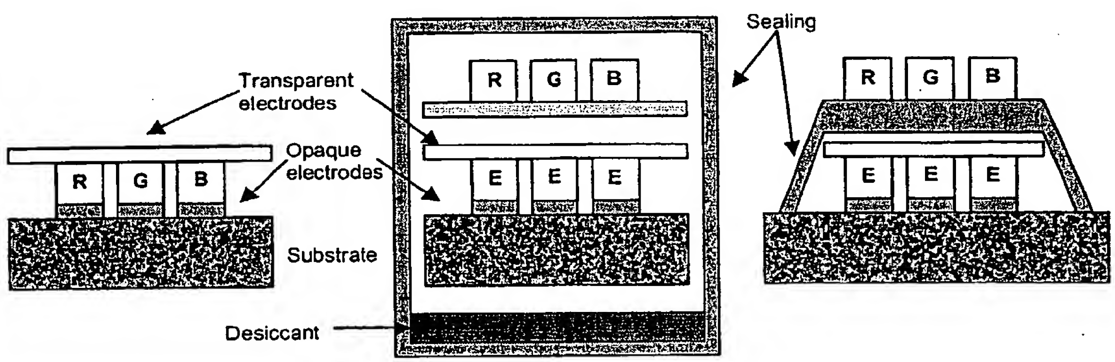
Exposure of the device to air results in the growth of black spots, which represent areas where the cathode[4] and the organic emitter are delaminated. Anode and hole transport layers do not seem to play any role in the formation of dark spots. Running the device in a nitrogen atmosphere does not lead to any growth of black spots[5]. However the number of black spots is predetermined by defects introduced during device production – the types of these defects are discussed in Ref. 5. Hence the sealing in general will only prevent the growth of these spots.

Based on several years of serious research and development efforts, over 10,000 hours half-life at 20mA/cm^2 (or over 100,000 hours at 100 Cd/m^2) are reported by different sources for properly produced and sealed devices[6,7,8,9], while the same devices typically live for a few 10 hours under unsealed ambient conditions. How to cost-effectively achieve conditions in low-cost production for the devices to reach storage lifetimes of more than 10 years has become a major issue for development in the OLED research and development. This report summarizes our contributions to that effort in the area of OLED-based displays.

2. EXPERIMENTAL

2.1 Considerations for Color Processing

The fabrication of triad pixel color displays requires the side-by-side patterning of red, green and blue sub-pixels (Fig. 1a). Since the OLEDs are extremely moisture sensitive, any kind of wet processing is incompatible if applied directly on the OLED. The use of shadow masks during the evaporation of organic materials to pattern the individual colors is not feasible for high resolution displays – hence an approach as sketched in Figure 1a cannot be used. In the case of active matrix OLEDs (AMOLEDs) on silicon[10] where the anode is the opaque bottom electrode (Fig. 3a), the obvious method is to pattern either color filters or color changing media (CCM) on a separate substrate (Fig. 1b), precisely align this substrate to the OLED device substrate and seal. This method, however, is quite difficult and expensive to implement, especially for high resolution microdisplays. Since this type of sealing can be made spacious, there is ample room for desiccant. To protect the OLED from damage during the wet CCM or color filter process, a proper alternative sealing procedure is shown in Figure 1 c which is applied immediately after deposition of the top electrode, allowing it to protect the OLED structure very efficiently. Therefore a preferred method will be to encapsulate the OLED device substrate with a suitable material so that the color forming materials can be directly patterned over it, using wet chemical processing.



(a) (b) (c)

Figure 1: Color OLED microdisplay sealing considerations: (a) individual emissive pixels (R,G,B) with the color of the light defined by the emitter itself, (b) the emitter (E) is either blue/ultraviolet and the color is achieved via color conversion media (R,G,B - CCM)[11] or the emitter is white and appropriate color filters (R,G,B) are used. The sealing approach in (c) protects the OLED structure under operation, during storage and during the production of the CCMs or the color filters.

2.2 Objectives

Key questions to be addressed when evaluating a proper sealing technique include:

- Is the sealing technique compatible with the OLEDs
- Do we achieve the required moisture and oxygen barriers
- Are the processing conditions destroying parts of the OLED structure
- Are the sealing-films transparent enough, are the OLED layers mechanically stable enough for the seal to be applied on top of them
- Is the wettability of the OLED top surface sufficient
- Is the OLED top surface planar enough
- How do the sealed devices compare to unsealed ones in their initial operation without being exposed to air or water
- Does the sealing provide the desired beneficial effects
- How does the chosen sealing method compare to alternate methods

These efforts were planned as a first-pass investigation into the viability of this sealing method.

2.3 Sealing Process

A few selected requirements for a sealing process for OLEDs are:

- Low permeability for oxygen and moisture (see [12])
- Compatibility with all device processing steps regarding:
- Materials properties
- Processing temperatures
- Thermal expansion coefficients
- In some cases the seal has to include a comparatively high volume of desiccant in order to trap the moisture penetrating the seal with time.
- The seal should not decrease the viewing quality of the display
- The seal has to withstand the processing conditions of the CCM or color filter production on top of it

Battelle Memorial Institute[13] has developed a new coating technology that renders polymer films impervious to oxygen and water vapor[12]. According to Ref. 12 the main reason for many potential OLED manufacturers to prefer glass over flexible substrates is the low permeability of glass. The present Battelle sealing technology claims to make polymer films virtually impermeable without changing their flexibility and is based on the application of an optically clear film. Battelle's technology relies on a vacuum-based process known as polymer multi-layer (PML) approach. All components are applied in a single processing sequence, avoiding the costly change of conditions other multi-layer approaches impose. Moreover the processing in vacuum ensures the highest level of cleanliness. The estimated permeation rate for the Battelle process for

oxygen is below 10^{-3} cubic centimeter/m^2/day[12]. The Battelle process applied to eMagin OLED test-structures is shown in Figure 2. The Battelle process has been licensed to Vitex Corporation for commercialization.

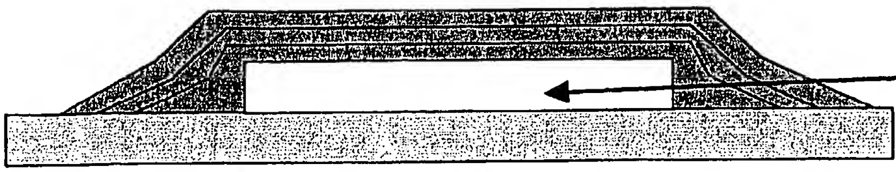


Figure 2: Barix™ sealing process applied to OLED test-structures of eMagin. The OLED stack, including the electrodes (white) is placed on a substrate (light gray) on top of which three sealing layers are applied: two so-called polymer multi-layer (PML) smoothing layers (dark gray) as well as a center layer consisting of a barrier oxide, nitride or oxynitride (vertical patterned layer).

2.4 Test Structures for the Sealing Process

The evaluation of processes, materials and device architectures for complex products like AMOLEDs often relies on the use of specifically designed test-structures which are closely related to the product but are significantly less expensive and less complex in production and handling. The test structure we have chosen is shown in Figure 3. In order to compare the results obtained in our test experiments with and without sealing after exposure to different external agents and after operation to literature data we chose a well known OLED composition (Figure 3). For OLEDs we defined the appropriate test-structure to be a semi-conventional up-emitting OLED. We used a silicon substrate covered with a high work function metal anode. On top of this electrode we deposited Copper Phthalocyanine (Cu-Pc) and N,N'-Di(napthalen-1-yl)-N,N'-diphenyl-benzidine (NPB) as well known efficient hole transport layers[14]. These layers are followed by either a doped host layer and a layer of tris(8-hydroxyquinoline) aluminum (Alq_3) or by an Alq_3 – layer only (see Table 1). Thin, semi-transparent films of Al-Li , Mg-Ag, and Ca all exhibit similar moisture sensitivity characteristics to the proprietary cathode used in this experiment. Standard device test structures were used for which a long internal historical data base exists, as opposed to our newer longer life, optimized devices.

TABLE 1: SUMMARY OF THE INVESTIGATED SAMPLES.

SAMPLE	TREATMENT
Doped Alq_3 – white emission	Tested immediately after production, unsealed (Figs. 3,4,5,6)
Doped Alq_3 – white emission	Sealed and tested immediately after sealing (Figs. 3,4,5,6)
Undoped Alq_3 – green emission	Tested immediately after production, unsealed (Figs. 3,7-10)
Undoped Alq_3 – green emission	Sealed, operated for 1 week in water, stored 1 month in air and tested (Figs. 3,7-10)

2.5 Test Methods

The last common processing step for all our test devices was the deposition of the top electrode. We have maintained a group of control devices which received no sealing and a group of devices which were covered as described in Section 2.3. Furthermore we also had unsealed control devices which went through all the transfers as well as the sealing conditions *without* actually receiving the seal. We have tested the as-prepared samples as well as their sealed counterparts in air, then exposed them to air and dipped them in water and also operated them under these conditions. Our test consists of measuring the electrical and optical parameters under operation with a fully automated computer-controlled combination of a Keithley

2400 source measure unit and a Spectrascan PR 650 Colorimeter. The previous experiments yield current voltage curves. Furthermore we took images of the samples under operation using a microscope equipped with digital imaging system. When mentioning test results and particular samples we always refer to experiments performed under identical conditions on several tens of active cells with active areas of 0.1 cm^2 each. We always report the *typical* result for the whole ensemble. The test method requires the device to run for a few seconds at typical current density of 10 mA/cm^2. Hence we are able to really monitor the "state" of the device at a given point in time, even if unsealed, since a few seconds of operation at these mild conditions does not change the device properties.

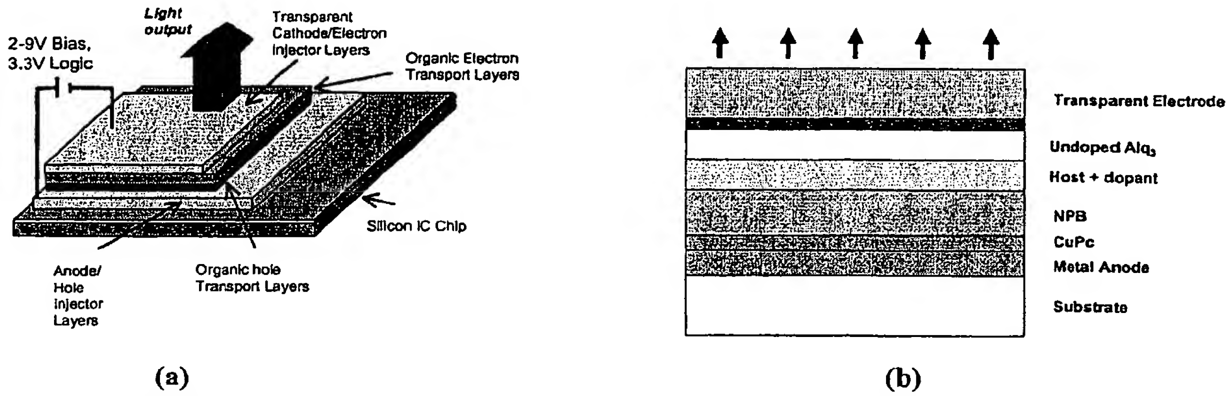


Figure 3: (a) Vertical arrangement of the planar layers required for the emissive elements of an AMOLED; (b) test structure, mimicking all the properties of (a) without using an actual silicon chip

3. RESULTS AND DISCUSSION

3.1 Sealing

The first question to be answered was the applicability of the sealing on our test structures. The adhesion of the barrier layer was observed to be very good on the thermal oxide (SiO_2) of the silicon substrates. It was checked with a standard cross-hatch pull test. For some metal oxide electrodes we observed an adhesion problem. Furthermore we observed that the sealing-films are definitely transparent enough, that the OLED layers are also mechanically stable enough for the seal to be applied on top of them and that the wettability of the OLED top surface is sufficient. Furthermore the OLED top surface seems also to be sufficiently planar. It has to be stressed that the sealing process has a lot of parameters that can be optimized to the particular object of sealing, e.g. the use of surfactant layers, which promote wettability of the surface to be coated.

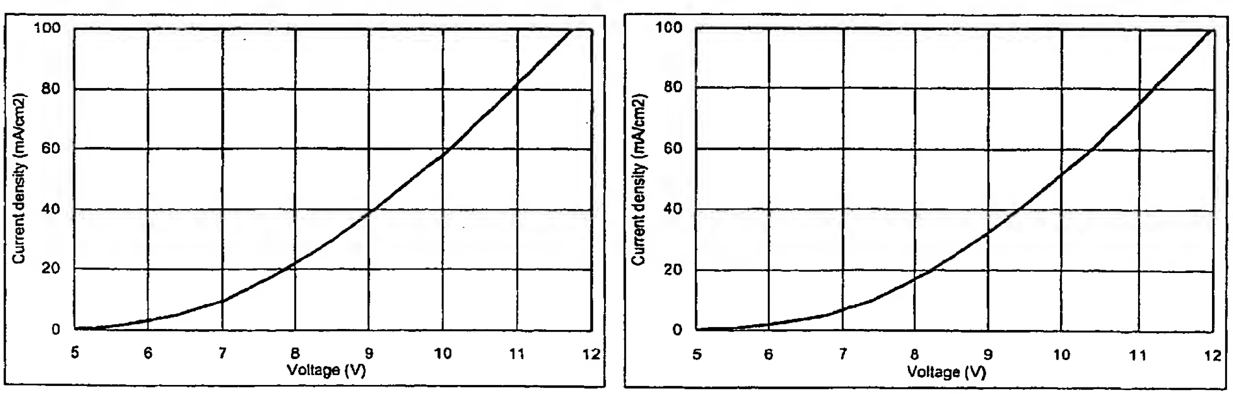


Figure 4: IV curve of the white emitting test structures before (left) and after (right) sealing.

3.2 Current Voltage (IV) Curves of White Emitting Test Devices Before/After PML Sealing

Applying the seal to the test structures did not result in any changes in the IV-curves of the devices as can be seen in Figure 4. Hence we conclude that the process is compatible concerning leaving the optoelectronic properties of our OLEDs unchanged.

3.3 Electroluminescence (EL) Spectra of White Emitting Test Devices Before/After PML Sealing

Applying the seal to the test structures did result in slight changes of the electroluminescence (EL) spectra. Since the CIE color coordinates remain unaltered the change is not a problematic one for the intended display application. We assume the change is due to modified conditions for the multiply reflected light in the complex organic/inorganic structure shown in Figure 3. Hence the sealing process is also compatible with the OLEDs concerning the spectrum of the emitted light.

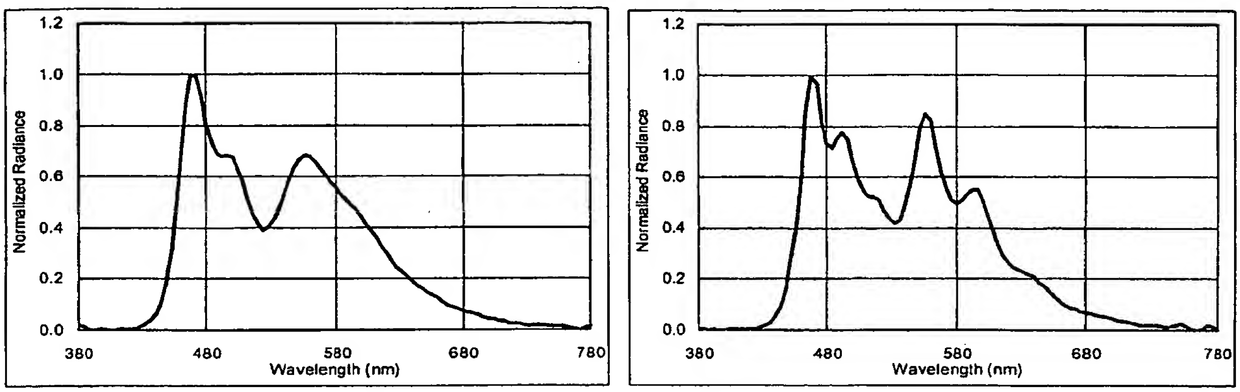


Figure 5: Normalized electroluminescence spectra of the white emitting test structures before (left) and after (right) sealing.

3.4 External Efficiency of the Electroluminescence of White Emitting Test Devices Before/After PML Sealing

Since we assume the change in the EL emission spectra after sealing to be due to modified conditions for the multiply reflected light in the complex organic/inorganic structure shown in Figure 3, we need to check whether the external efficiency of the test structures had changed, i.e. whether we loose or gain light intensity due to the introduction of the sealing layers. One of the possible ways to do this is to measure the power efficiency of the devices prior to and after sealing. Apparently applying the seal to the test structures did result in a slight increase of power efficiency as depicted in Figure 6. Hence the sealing process is also compatible with the OLEDs concerning the efficiency of the EL. After ensuring the compatibility of the sealing process with the test structures it is important to check how effective the seal is as a moisture and oxygen barrier.

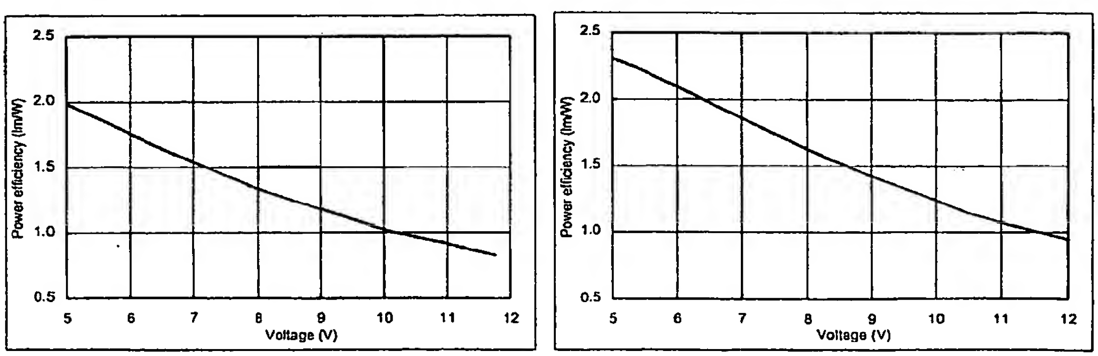


Figure 6: Power efficiency of the EL of the white emitting test structures before before (left) and after (right) sealing.

3.5 Current Voltage (IV) Curves of Green Devices Before/After Operation in Water and Storage in Air

We have investigated several driving and storage conditions in air and water and show the complete set of data for one of these conditions: brief storage in air after sealing, one week of continuous operation in water with a constant current density of 20 mA/cm^2 and finally one month storage in air. Figure 7 shows the IV curves of a test structure before sealing as well as after sealing and operation/storage under the described conditions. It is evident that the IV curve shows a slight change after operation/storage with the achieved current at constant voltage of operation dropping to approximately 80 % of its initial value. Small changes in operation parameters are not uncommon in either organic or inorganic semiconductor devices due to the fact that devices immediately after production are not necessarily in their stable form. This device characteristic is even less noticeable in newer versions of our white OLED devices, but is characterized in the standard device structures well enough to permit differentiation form sealing or moisture/oxygen permeation effects. Hence burn-in procedures or annealing steps are used to let devices undergo stabilization before they leave the processing area. Since these burn-in procedures might complicate the interpretation of our test results we have refrained from them and therefore the operation in water reveals a change in the IV-curve of the test structure which is not unexpected and also commonly observed in lifetime tests of other sealed organic devices.

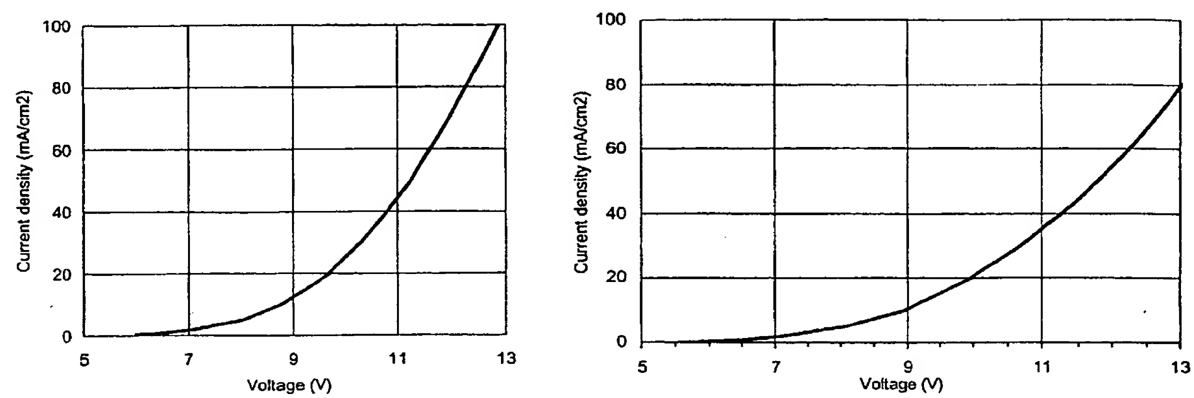


Figure 7: IV of the green emitting test structures before (left) and after (right) sealing + 1 week of operation in water + one month storage in air.

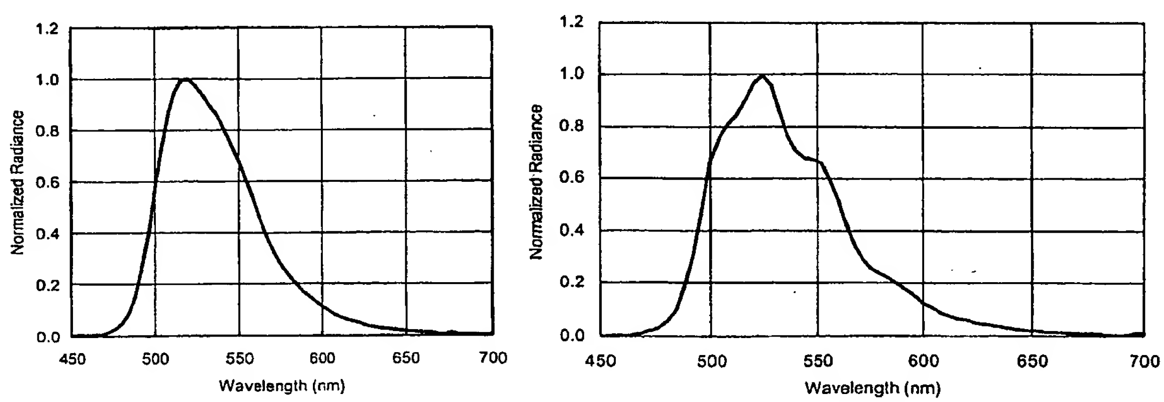


Figure 8: Normalized EL-spectra before (left) and after (right) sealing + 1 week of operation in water + one month storage in air.

3.6 EL Spectra of Green Devices Before/After Operation in Water and Storage in Air

The change in the EL-spectra is mainly due to the sealing process itself since we compare the performance of the unsealed and the sealed + operated + stored device. The results, reported in Figure 5, strongly support this conclusion.

3.7 External Efficiencies of Green Devices Before/After Operation in Water and Storage in Air

A similar efficiency increase as observed in Figure 9 is also reported in Figure 6. This leads to the conclusion that not the operation and storage leads to the increase in efficiency, but that the sealing layer might work as an effective antireflection coating on an OLED test structures. In this context the IV curves are the only ones not affected by the optical properties of the sealing layer while both power efficiency as well as spectrum will be influenced by changes in the OLED optics introduced by the sealing layer. In fact the external power efficiency is derived from the spectrum, the determined angular distribution of the emitted light and the electrical data – hence it contains the combined information of the optical and the electrical test. Therefore the change in emission spectra (Figs. 5/8) and the apparent increase in efficiency are both consistent with the assumed antireflection effect of the sealing layer. We have not conducted a quantitative analysis at this point but the expected refractive indices of the top OLED electrode layer and the sealing material are in agreement with this explanation.

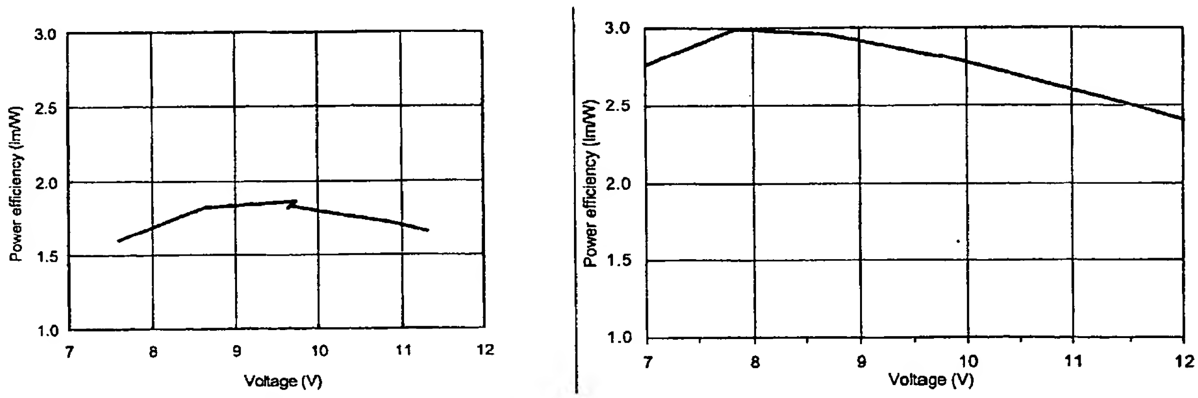


Figure 9: Power efficiencies before (left) and after (right) sealing + 1 week of operation in water + one month storage in air.

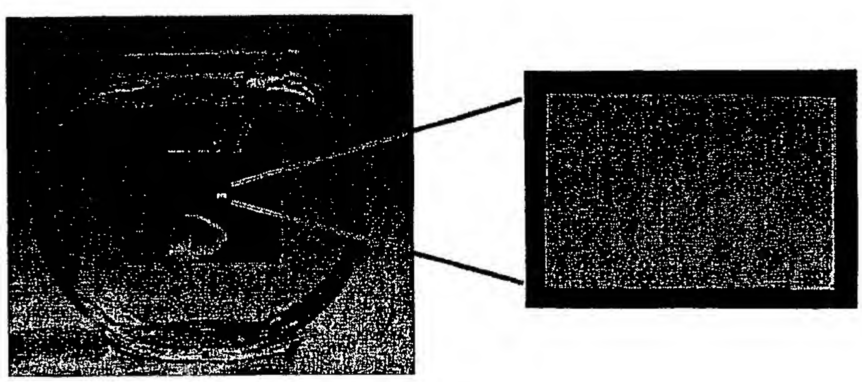


Figure 10: Image of the operating Alq_3 devices after sealing + 1 week of operation in water + one month storage in air. The active area is approximately 2.5 x 4 mm in size (fishbowl image courtesy of Battelle)

The effect of operation under water on the sealed structures is shown in Figure 10. The observed black spot pattern is well in agreement with published literature on the subject[4,5]. An unsealed test structure with transparent electrodes does not function anymore after more than 1 week of operation in water plus one month storage in air. Conventional sealing techniques lead to similar black spot images under the given conditions. Hence we conclude from both the operational characteristics as well as the low amount of dark spots that the Battelle process provides a reduction in size and rate of formation of black spots.

4. CONCLUSIONS

We have shown that the PML sealing process developed by Battelle is compatible with all steps of the OLED-based microdisplay production.. Our results document that encapsulating OLEDs with the conformal polymer/metal(oxide)/polymer multi-layer technology leads to significantly improved stability, both under operation as well as for storage purposes in comparison to unsealed displays.

5. ACKNOWLEDGEMENTS

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